

Assessment of Mercury Environmental Fate and Transport from INEEL Waste Processing Facilities for Long-Term Stewardship Concerns and Development of Improved Modeling Methods

Understanding the Movement of Mercury in the Environment Surrounding the INEEL

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SUMMARY

Environmental fate and transport of the toxic air pollutant mercury (Hg) is currently a high-priority regional concern for the INEEL, and national and global concern for the U.S. Environmental Protection Agency (EPA). At the INEEL's Idaho Nuclear Technology and Engineering Center (INTEC), significant quantities (est. 40 kg/year) of Hg may have been released over 37 years of Environmental Management's (EM's) high-level waste (HLW) calcination operations. The EPA is very concerned about the continued global buildup of Hg in the atmosphere and aquatic ecosystems, and has recently invested heavily in Hg research to better understand its complex environmental cycling.^{1,2} This task is a joint INEEL and U.S. Geological Survey field research effort to better understand the fate of Hg emissions (operational component) from the INEEL's calciner (New Waste Calcining Facility [NWCF]) and contribute at a national level to the scientific understanding of local, regional, and global Hg fate and transport (research component). This task consists largely of Hg fallout sampling in precipitation and soils in two geographic areas: the INEEL and surrounding airshed, which includes the Eastern Snake River Plain and adjacent elevated areas; and two high-elevation, mid-latitude glaciers—the Upper Fremont Glacier (UFG) in Wyoming and the Inilchek Glacier in Central Asia. Glacial snowpack provides a preserved annual record of both regional and global Hg fallout that can be used to answer questions about long-range INEEL transport and help to fill existing data gaps on the relative importance of local versus global source contributions to fallout.

Samples of snow ($n = 130$), rain ($n = 6$), soil ($n = 103$), and air ($n = 5$) were successfully collected on the INEEL and at 11 regional background sites from January through March. Analytical results for snow before startup of the calciner indicated relatively low Hg concentrations within the range of the western regional background (3 ng/L) and unexpectedly high concentrations (10 to 20 ng/L) near INTEC, which may be due to Hg evasion from the soil followed by entrapment in the overlying snowpack. Because startup of the NWCF was delayed until March 15, 2000, and no appreciable snow fell after this date, snow sampling to determine NWCF fallout rates on the INEEL was not possible. Snow samples were successfully obtained from the UFG in May ($n = 20$) and the Inilchek in July ($n = 50$). The UFG data indicated very low (0.3 to 0.9 ng/L) concentrations relative to regional background, and the Inilchek samples have not yet been analyzed. Surface soil sampling around INTEC showed (a) very low Hg concentrations overall compared to background levels in similar soils across the U.S., (b) a 4% per cm reduction in Hg concentration with depth, and (c) significantly higher Hg concentrations in soils under shrubs (likely due to fixation by organic matter) and in depressions (likely due to Hg runoff). Using these data, mass balance calculations were able to account for approximately 40–300 kg of calciner Hg in the soil, which is only 3–20% of the estimated total Hg emitted over the 37-year calciner operating history. These results provide evidence that much of the Hg deposited from calciner operations may have been reduced in the soil and reemitted as Hg(0) to the global atmospheric pool.

TASK DESCRIPTION

Summary of Sampling Activities

Table 1 summarizes the multimedia sampling activities during FY 2000. All samples were analyzed for total mercury (THg), and a small fraction were analyzed for methylmercury (MHg). Total Hg includes all chemical forms of Hg, including (a) divalent Hg(II), which readily deposits and is the predominant form of Hg emitted by the calciner; (b) elemental Hg(0), which comprises the bulk of the global atmospheric pool, but does not readily deposit (until some of it is oxidized by downwind atmospheric chemistry); and (c) methyl mercury, which is of concern because it is a more toxic species and bioaccumulates in aquatic organisms (up to 10^7 times the water concentration). Snow and rain were sampled because these are the dominant scavenging mechanisms that transport Hg from the atmosphere to the ground, where it is of primary concern (see Reference 1). Also, precipitation concentrations can be linked with precipitation time intervals to calculate Hg deposition rates, which can then be used to validate deposition models. Snow was the primary sampling medium because it is the dominant form of precipitation in this region and because it usually remains on the ground, allowing sampling at a later date. However, because startup of the NWCF was delayed until March 15, 2000, and very little snow fell on the INEEL after this date, only five snow samples on the INEEL were obtained during NWCF operations. Because of the lack of the INEEL NWCF data, proposed source apportionment methods (principal component analysis and isotopic ratios) were not implemented. Limited rain sampling was done after this time because of the continuing dry conditions this spring and the difficulty of coordinating rain sampler deployment during the few short-term rainfall events that occurred. Soil sampling was successfully

Table 1. Summary of mercury sampling activities for FY 2000.

Location	Month/00	No. Samples	Analyzed?	Purpose
<u>Snow</u>				
INTEC 5-km Grid	Jan 4–6	50	Yes	Pre-NWCF startup baseline
(Figure 1)	Mar 17	8	Yes	Post-NWCF impacts
Background sites (10)	Dec 20	15	Yes	Regional background surrounding
(Figure 2)	Jan 17–19	24	Yes	INEEL
	Mar 25–26	34	Yes	
W. Slope Teton Range (5 sites from 6,000 to 10,000 ft)	Jan 17–18	20	Yes	1) High precipitation/fallout rates 2) Sensitive “downwind” ecosystem
Upper Fremont Glacier, WY (13,200 ft)	May 1–8	20	Yes	1) Long-range INEEL transport 2) preserved annual fallout record in snowpack
Inilchek Glacier, Asia (16,000 ft)	July	50	No	Global fate/transport data gaps
<u>Rain</u>				
INTEC Grid – NE1, NE2, NW1	May	6	Yes	1) Rain sampling method 2) NWCF fallout
<u>Air</u>				
INTEC Grid—1–4 km downwind of NWCF plume; rest area	Mar	5	Yes	1) Air sampling method, 2) regional background
<u>Soil</u>				
Experiment 1	Jul	24	Yes	Sampling methods-depth
Experiment 2	Jul	20	Yes	Sampling methods-vegetation
INTEC Grid	Aug	59	No	Residual cumulative deposition

accomplished in late summer to determine the Hg variability in soil (Experiments 1 and 2) and to estimate the cumulative residual Hg load in the soil around INTEC after 36 years of calcining operations.

Most of the sampling on the INEEL was conducted on the INTEC 5-km grid—an array of 64 sampling locations located on 22.5-degree radials at distances of 1, 2, 3, and 5 km (Figure 1). This grid was designed, based on previous air modeling studies of the INTEC 250-ft main stack,³ to provide an adequate assessment of the fallout pattern around the facility with a reasonable number of samples. The 10 background sites (Figure 2) were selected based on their location surrounding the INEEL, their likelihood of snowfall accumulation, and winter access. Snow was also sampled in January on an elevation gradient extending from the Teton Valley outside Driggs, Idaho (6,000 ft) to the summit of Grand Targhee Ski Area (10,000 ft). This area was selected because it (a) receives very high snowfall (maximizes air pollutant scavenging) and snow accumulation (allows sampling of a long-term fallout period), (b) is an optimum location to investigate changes in Hg fallout with elevation (current high interest to EPA), and (c) is located between the INEEL and concerned public interest groups in Jackson, Wyoming (stakeholder considerations). Snow sampling was conducted at the Upper Fremont Glacier (UFG) and Inilchek Glacier sites because of (a) the comprehensive annual fallout record preserved in their winter snowpack, (b) their mid-latitude locations and elevations, and (c) ongoing research activities at these locations by the U.S. Geological Survey, which allowed sharing of field trip expenses. The UFG site was also investigated to determine if long-range transport of Hg from the INEEL could be detected.

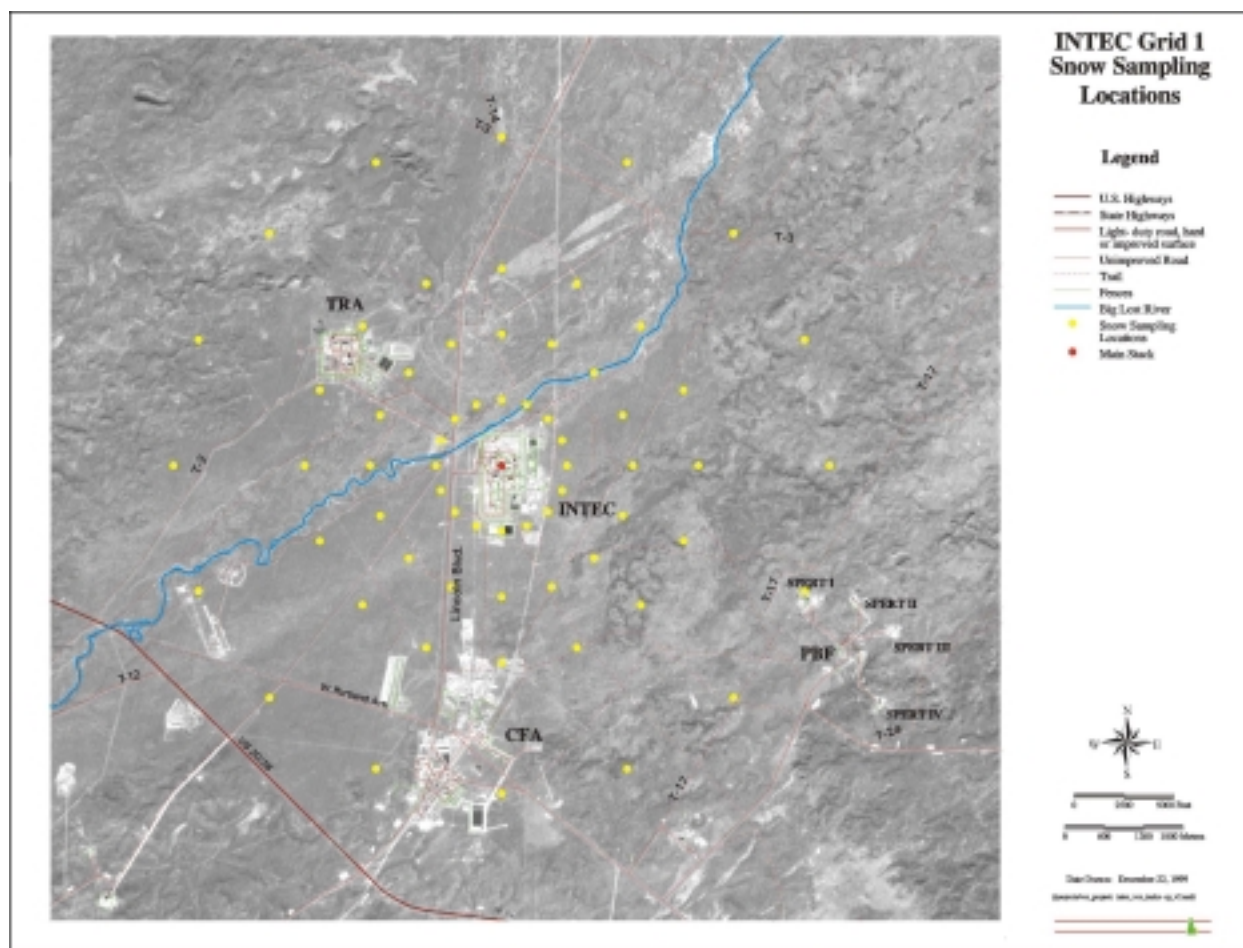


Figure 1. INTEC 5-km sampling grid—64 locations on 22.5° radials at distances of 1, 2, 3, and 5 km.

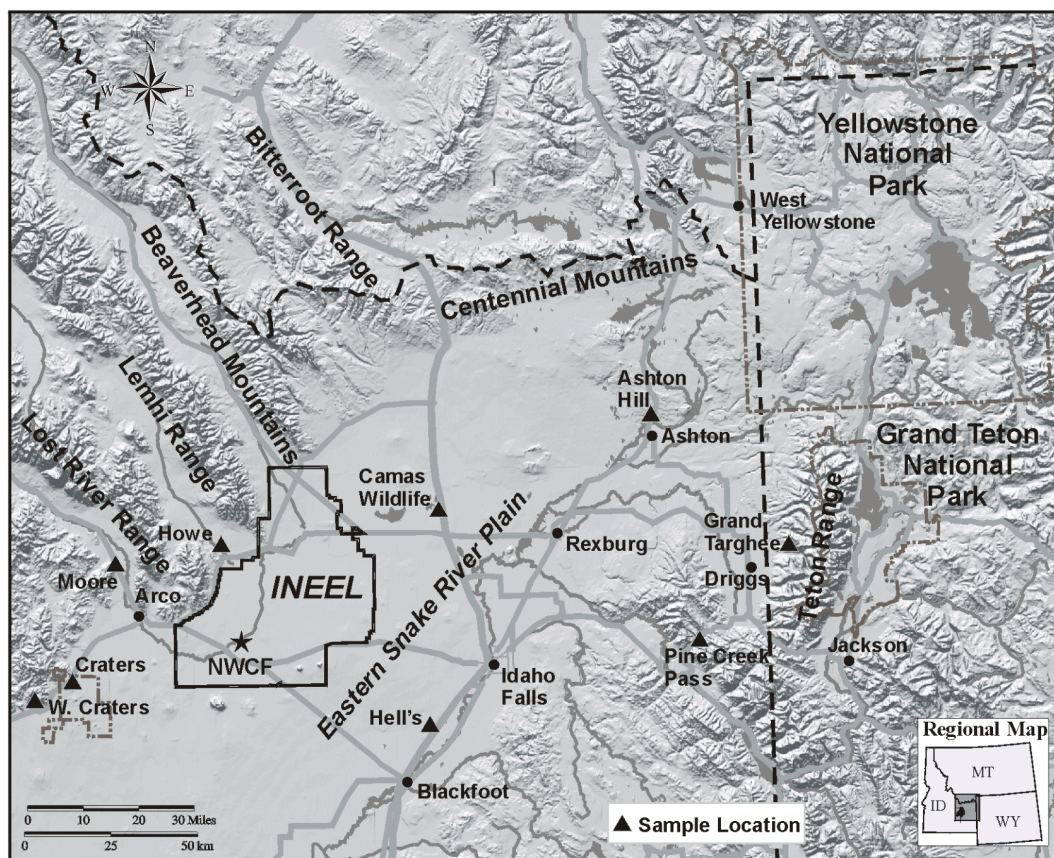


Figure 2. Regional background snow sampling locations (Pebble Creek site is 10 km south of map).

Sampling and Analytical Methods

Laboratory analysis and sample bottles were provided by the U.S. Geological Survey Wisconsin District Mercury Laboratory (WDML) in Madison, Wisconsin. The WDML is one of two full-service environmental mercury laboratories in the U.S. All sampling is done using laboratory cleaned, Hg-free sampling bottles (30 mL, 250 mL, 500 mL, and 2 L) and shoulder-length gloves; the sample bottles are kept upwind during sampling.

For snow samples, the snowpack was excavated to the ground and the snow face was cleaned using a clean lexan shovel (Hg sticks to metal). At the INEEL, composite samples of the entire snowpack thickness (typically about 20 cm) were collected using special precleaned 500 mL and 2 L teflon bottles. At background and glacier sites with significant (>50 cm) snowpack, a snow pit was dug, and 10-cm interval samples were collected in addition to a composite. Snow density measurements were made which are later multiplied by the snow depth to determine the snow water equivalent (SWE) at each location (SWE is equivalent to the mass [kg] or depth of water [mm] per unit area). After sampling, the bottles were double-bagged, placed into a cooler, and shipped frozen to the WDML.

Rain samples were collected in portable collectors consisting of custom-made 20 L teflon bag liners inside 55-cm-wide × 37-cm-long × 15-cm-high plastic storage containers (Sterilite® Clearview). The collectors were deployed at model-predicted maximum impact downwind locations on the INTEC

Grid. After the rainfall event (typically overnight), the sample (if any) was poured into a 500 mL teflon bottle. Soil samples were collected in 30 mL teflon vials after surface litter and pebbles were removed.

In the laboratory, samples were melted, acidified (to keep the mercury oxidized and in solution), and analyzed for total mercury (THg) and methylmercury (MHg).⁴ THg includes all chemical forms of Hg, including divalent Hg(II), elemental Hg(0), and methyl mercury. THg analysis was performed using EPA Method 1631, “Mercury in Water by Oxidation, Purge and Trap, and Cold Vapor Atomic Fluorescence Spectrometry (CVAFS),” with modifications. MHg analyses were performed using EPA Method 1630, “Methyl Mercury in Water by Distillation, Aqueous Ethylation, Purge and Trap, and CVAFS,” with minor modifications. For some samples, a filtered/unfiltered (FTHg/UTHg) split was analyzed, which can provide information on whether the Hg was associated with particulate or existed as a gas in the atmosphere (which provides evidence of the fallout source). Soil samples are analyzed in a similar way, except that a subsample was first taken and then digested in a mixture of nitric and sulfuric acid. The laboratory uses a rigorous QA/QC procedure, which includes duplicate analyses on every sample, spike recovery analyses at least once every 10 samples, quality control check samples, and bubbler blanks to check for background contamination (see Reference 1).

Sampling Results—Snow

A summary of the Hg snow sampling results (in ng/L) for the INEEL and background sites is provided in Table 2. For comparison, the EPA maximum contaminant level (MCL) for Hg in drinking water is 2,000 ng/L. Most of the snow samples were taken before the NWCF was started on March 15, 2000. A few background sites with late winter snowpacks (Pebble Creek, Pine Creek Pass, and Ashton Hill) were sampled after the startup date, but very limited snow sampling was possible on the INTEC grid because of the late calciner start date.

The INTEC grid sampling results conducted before calciner startup showed statistically higher Hg concentrations compared to the background sites, which was very surprising given the NWCF had not operated through the winter (see Figure 3). The following are possible reasons for these unusual results:

- Another unknown Hg source at INTEC operating during the period of snowfall (primarily in late December 1999) before the sampling (Jan 4–6, 2000)—investigated but none found. The previous calciner campaign (NWCF4) was completed in May 1999, and the High-Level Liquid Waste Evaporator was last run in July 1998.
- Soil cross-contamination during sampling—not likely because (a) the increase in concentration around INTEC was relatively uniform, which would require consistent cross-contamination with soil, and (b) the “ultraclean” sampling techniques specifically avoid contact of any sampling material with the soil surface; these methods have been previously used numerous times without any indication of cross-contamination.
- Reemission of deposited Hg in the soils around INTEC followed by absorption or hold-up in the snowpack prior to release to the atmosphere. This is a likely explanation because (a) the filtered versus unfiltered fraction (FTHg/UTHg) for the January grid samples (0.42) were higher than what has been previously measured suggesting a gaseous Hg source, (b) a mass balance calculation on the soil sampling results (see below) indicates significant reemission loss of deposited Hg from soils, and (c) soil reduction of the deposited Hg(II) and reemission as Hg(0) has been well documented, especially in high pH, low organic content soils characteristic of the INEEL. Snow and soil flux experiments are planned for FY 2001 to confirm and quantify these reemission rates.

Table 2. Summary of Hg concentrations in snow (F/U = filtered/unfiltered; M/THg = methyl/total).

Sampling Location	No. samples	THg Concentration in Snow (ng/L)			Fraction F/U THg	Fraction M/T Hg
		High	Low	Avg.		
INTEC Grid (1/4–1/6/00, prior to calciner start):						
1 km radius	8	22	8.9	15	0.41	0.017
2 km radius	16	15	5.5	9.3	0.45	0.019
3 km radius	13	14	5.6	8.9	—	—
4 km radius	12	11	4.7	7.7	0.38	0.013
Average =				10	0.42	0.02
INTEC Grid (3/17/00, after calciner start):						
1 km radius	5	141	6.4	55	—	—
Background Sites [(A) = after calciner startup]:						
Pebble Creek	2	4.5	3.6	4.0	0.25	0.03
Pebble Creek (A)	3	2.7	1.1	2.1	.35	0.02
Hell's Half Acre	2	8.5	5.2	6.9	0.19	0.01
Pine Creek Pass	13	6.8	1.5	2.2	0.33	0.03
Pine Creek Pass (A)	6	5.9	1.7	2.2	0.26	0.01
Ashton Hill (top)	18	12	1.8	3.3	0.42	0.02
Ashton Hill (A)	5	5.0	2.3	3.8	0.21	0.04
Camas Wildlife Refuge	5	3.4	1.6	2.3	0.30	0.01
Howe area	3	6.2	1.2	3.8	0.38	0.01
Moore area	8	7.3	0.9	3.0	0.29	0.01
Craters of the Moon	7	3.7	1.9	2.6	0.37	0.01
West Craters	3	4.6	2.7	3.3	0.27	0.01
Average =				3.3	0.30	0.02
W. Slope Teton Range:						
Tetons – 10,000 ft	6	7.8	2.56	3.5	0.31	0.004
Tetons – 8,000 ft	4	9.3	3.6	4.0	0.28	0.04
Tetons – 7,000 ft	3	3.1	1.7	1.7	0.33	0.01
Tetons – 6,600 ft	4	2.6	2.0	2.6	0.19	0.01
Teton Valley	3	6.9	2.5	4.6	0.26	0.02
Average =				3.3	0.28	0.02

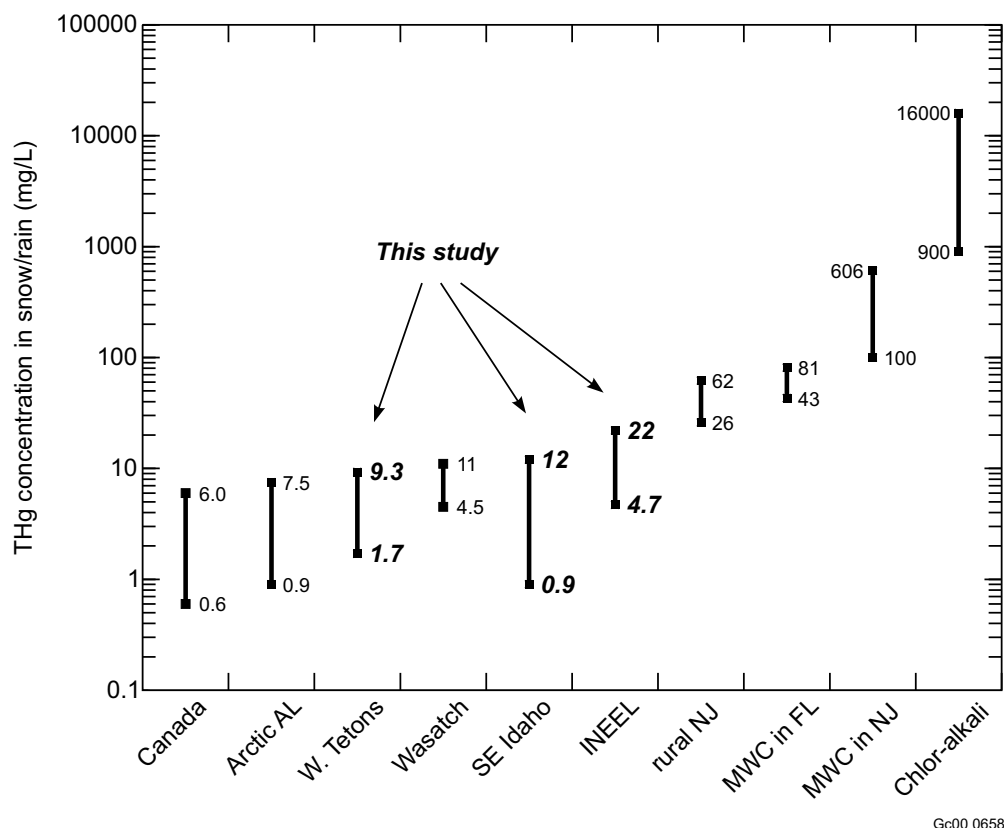


Figure 3. Comparison of snow THg concentrations found in this study (INEEL, SE Idaho, and W. slope Teton Range) with published values for precipitation (snow and rain) in other areas of the U.S. and Canada.

The INTEC grid snow sampling conducted after calciner startup indicated much higher concentrations at two locations (134 ng/L at 1 km northeast of INTEC and 141 ng/L at 1 km east-northeast). However, only a limited number of samples could be taken because of the poor snow conditions at this late date, and these samples may have been cross-contaminated with soil because of the difficulty of sampling the shallow (<2 cm) snow depth.

At background sites and the west slope of the Teton Range, snow concentrations were low (3.3 ng/L), similar to those measured in other remote low-impact locations, including arctic Alaska (0.9 to 7.5 ng/L),⁵ Ontario, Canada (0.6 to 6.0 ng/L),⁶ and in Utah's Wasatch Range (4.5 to 11 ng/L)⁷ (see Figure 4). Concentrations in precipitation in eastern industrialized areas have been measured at much higher levels, i.e., 26 to 62 ng/L in a rural New Jersey area,⁸ 606 ng/L near a municipal waste combustor (MWC) in New Jersey, and 16,000 ng/L near a chlor-alkali plant in an urban residential area.⁹

Snow sampling on the Upper Fremont Glacier, Wind River Range, Wyoming was successfully completed May 1–8, 2000. Twenty 500-mL snow samples were collected from the full winter snowpack (two replicate pits 2.3 m deep) on the glacier at 13,200-ft elevation. The analytical results indicated very low mercury concentrations (0.3 to 0.9 ng/L) relative to the expected background concentration in precipitation in this region (1 to 3 ng/L). The laboratory analytical QA procedures were verified, and the results were found to be valid. These low results are currently being investigated but may be due to excessive Hg volatilization in the snowpack driven by the unusually dry winter (and therefore relatively old age of the snow sampled) and the high solar radiation conditions that existed during the months prior to sampling (a reemission driver).

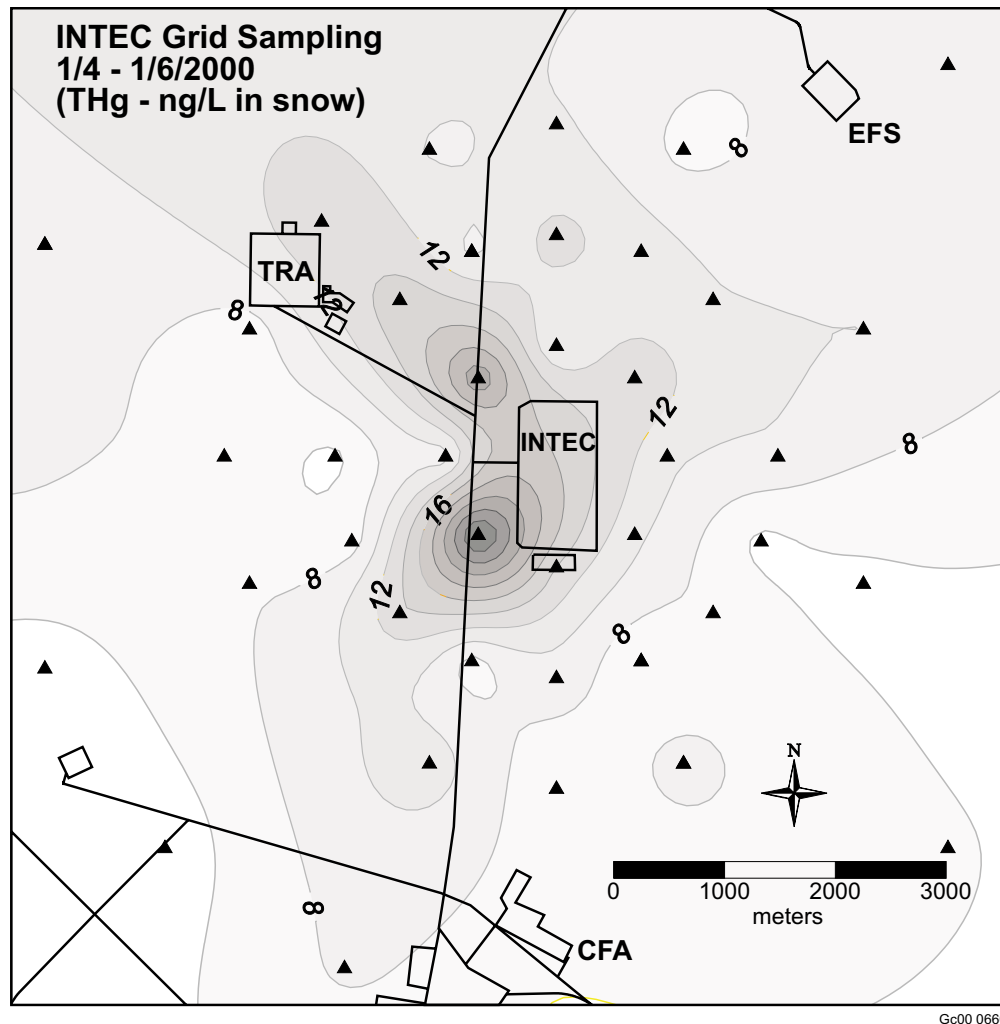


Figure 4. THg concentrations in snow (ng/L) sampled on the INTEC 5-km grid prior to startup of the calciner (3/15/00).

Sampling Results—Rain

Snow sampling on the Inilchek Glacier in Central Asia was successfully accomplished during the first two weeks of July 2000 in collaboration with a USGS glacier ice-coring expedition. Fifty snow samples from three 3-m-deep snow pits (two to three seasons of snow) were obtained and are currently being analyzed. These samples will help fill important data-gaps on global cycling of Hg in remote locations. The EPA Office of International Activities expressed interest in this work during a May 25, 2000 conference call.

Only three significant rain events occurred before shutdown of the calciner at the end of May 2000. Six rain samples were successfully obtained at three locations on the INTEC grid during one of these events (May 17, 2000). Two of these locations were chosen to be directly in the plume path (northeast at 1 and 2 km) and one was taken in the cross-wind direction (northwest at 1 km). Concentrations ranged from 14 to 20 ng/L in the plume path to 11 ng/L in the cross-wind direction. The demonstration of a successful collector design that is inexpensive and easily deployed is considered the most important result from these sampling activities.

Sampling Results—Air

Some limited air sampling was performed on the INTEC 5-km grid while the calciner was operating using a Bios AirPro portable air sampler (1 L/min) and goldtraps (gold-coated silica sand in a glass tube). The results ($n = 3$, $u = 2.23 \text{ ng/m}^3$) were within the global background range (1 to 3 ng/m^3) expected for this region. These results are not unexpected since goldtraps only trap elemental Hg(0), and emissions from the calciner are believed to be primarily in the divalent Hg(II) state.

Passive integrated mercury samplers (PIMs), still in a developmental stage, were obtained without cost through collaboration with the USGS Columbia Environmental Research Center (CERC) in Columbia, Missouri. Three of these units were deployed for 25 days on the INTEC sampling grid during calciner operations. Analytical results showed a blank contamination problem (from the stainless-steel shipping cans), and the results were not considered valid. The CERC has fixed the problem and may provide more PIMs (in different Hg-free shipping containers) for INEEL deployment in the future.

Sampling Results—Soil

Before comprehensive INTEC grid soil sampling, two experiments were conducted in July 2000 to determine Hg distribution in the soil around INTEC. The first experiment examined Hg concentration as a function of soil depth at a high-fallout location 1 km northeast of INTEC. Twenty-four soil samples were taken at 1-in. depth intervals down to 4 in. using a custom-made soil horizon sampler (borrowed from the State of Idaho INEEL Oversight Program). Results showed that Hg soil concentrations decrease about 10%/in. of soil depth (see Figure 5) and that overall soil concentrations near INTEC are low relative to background levels published for similar soils in other regions in the U.S. (50 to 60 ng/g).

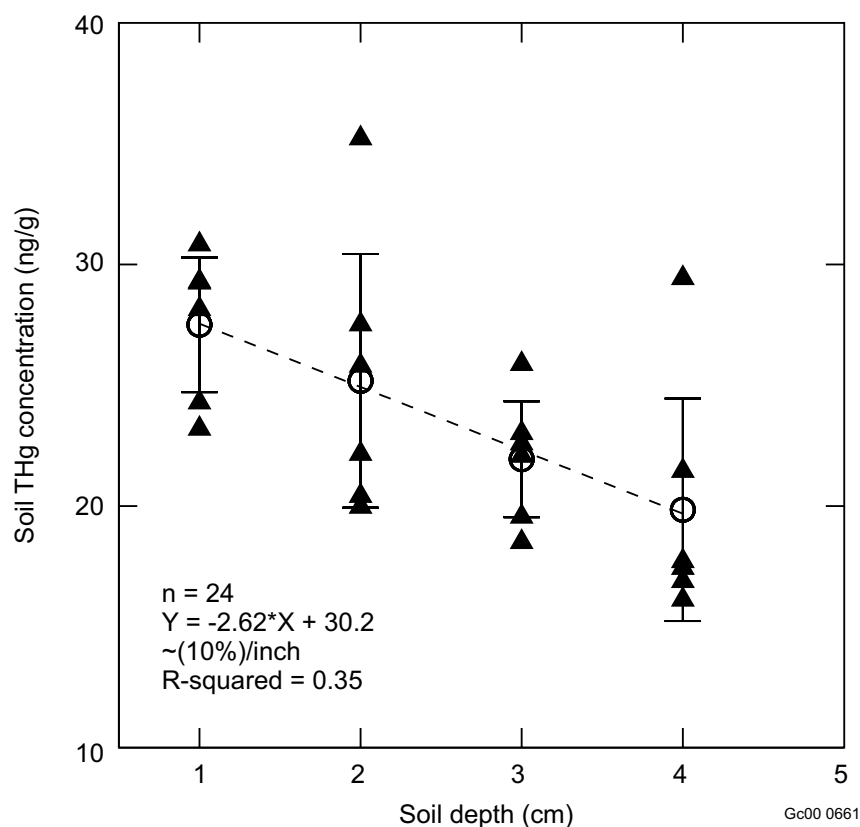


Figure 5. THg concentrations in soil near INTEC decreased approximately 10%/in. with depth.

The second experiment examined the variability of surface soil Hg concentrations as a function of vegetation type/cover and soil runoff features (rise vs. depression). These results (see Figure 6) show (a) Hg soil concentrations under shrub (sagebrush and rabbitbrush) canopies are a factor of 2 higher ($p < 0.05$) than soil concentrations in grassy or bare areas, likely due to fixation by the higher organic matter content of soils there, and (b) Hg soil concentrations in depressions are a factor of 2 higher than in adjacent elevated areas, likely due to runoff.

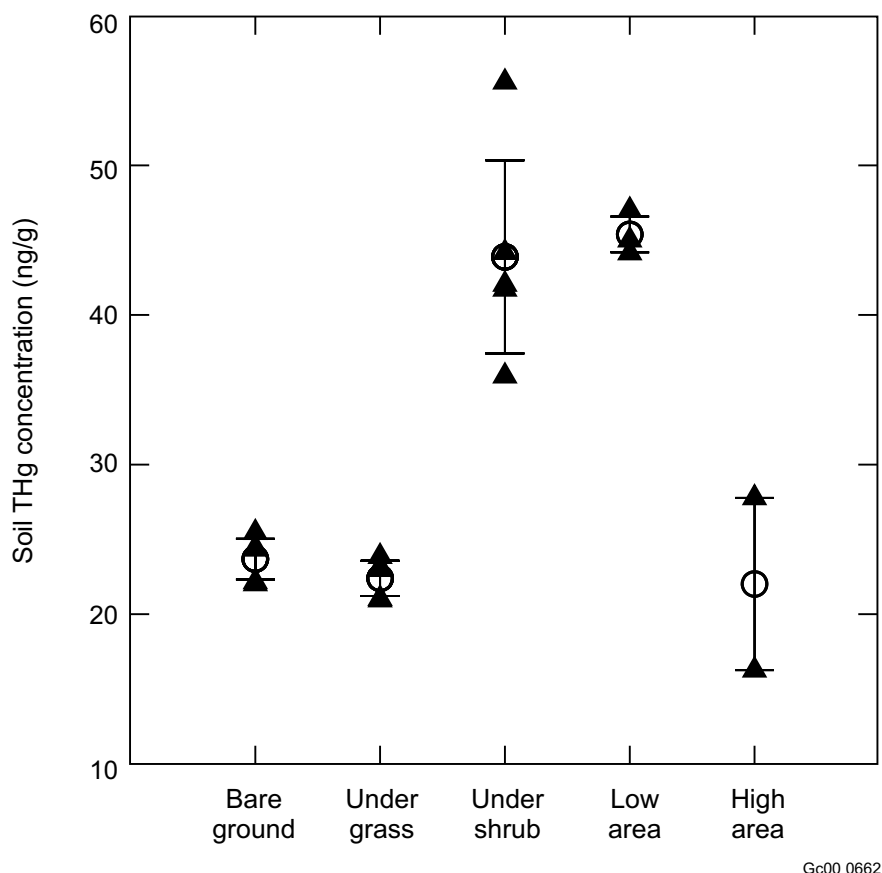


Figure 6. THg concentrations in soil under shrubs and in depressions were found to be a factor of 2 higher ($p < 0.01$) than concentrations in soil under grass, with no vegetation, or in elevated areas.

Comprehensive soil sampling results on the INTEC grid (see Figure 7) showed very low Hg concentrations overall compared to similar soils across the U.S. and a slight increase ($p < 0.05$) in concentrations within 5 km of INTEC (the “halo” in Figure 7). An estimate of the total calciner Hg load in the soil around INTEC and a mass balance of calciner Hg emissions over its 37-year operating history was made by (a) integrating surface soil concentrations across the 5-km grid (where most of the fallout was observed) and out to a conservative range of 20 km using Surfer[®] 7^a kriging and volume tools and a conservatively-low background level of 7 ng/g; (b) repeating this process at 1-cm depth layers, reduced in concentration by 4% per cm (from the soil depth experiment), down to “0” integrated concentration; and (c) summing the calculated Hg mass in the 1-cm layers. The results show a total surface soil load near the facility of 37–300 kg which is a small fraction (3–20%) of the estimated cumulative 37-year emissions from the calciner (1,500 kg).

a. Golden Software, Inc., Golden, Colorado 80401; www.goldensoftware.com

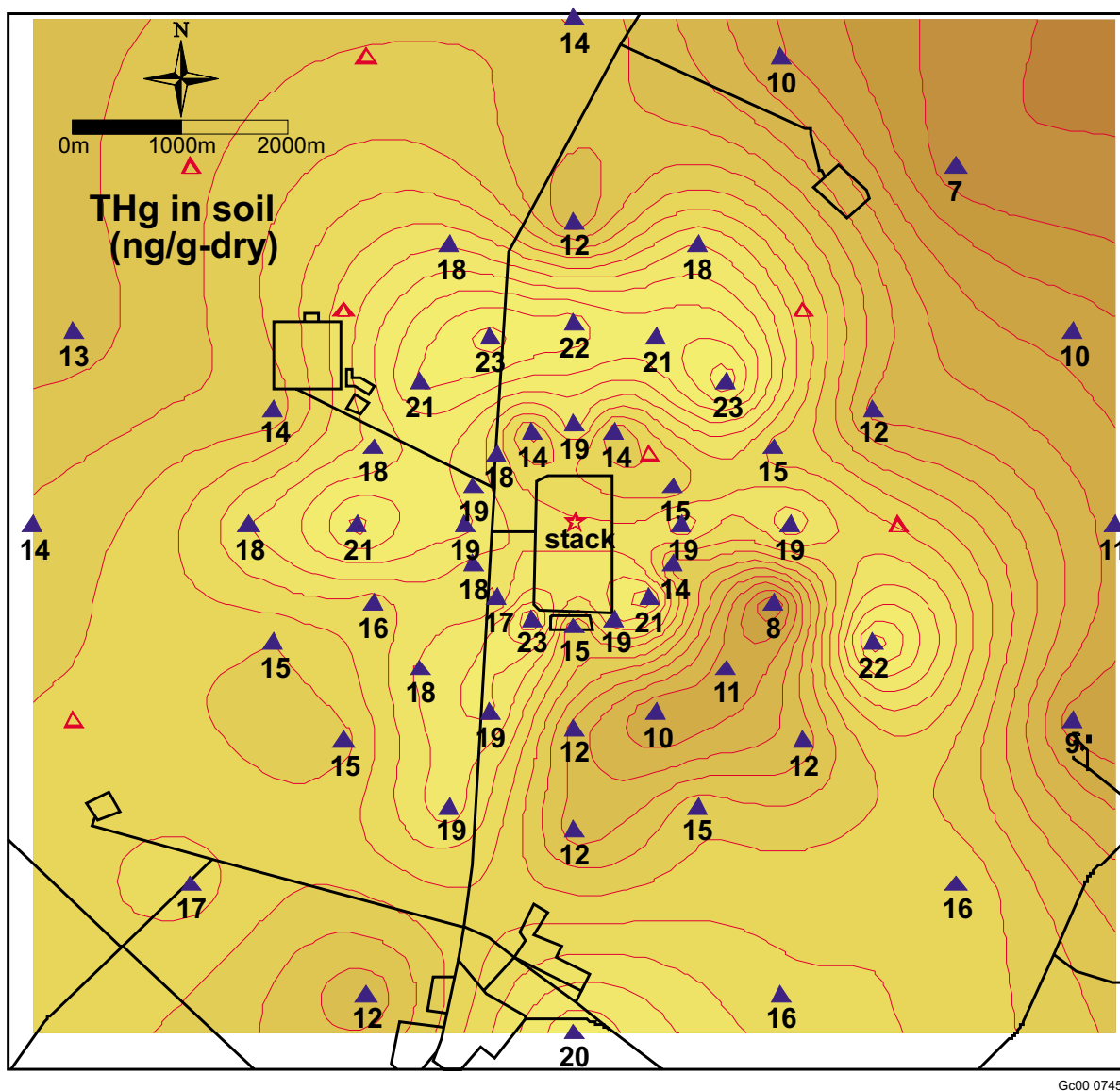


Figure 7. Hg concentrations (ng/g) in surface soil within 5-m of the INTEC main stack.

These soil sampling results, the mass balance calculations, and the high grid snow sampling results before calciner startup provide good evidence that most of the Hg initially deposited on the INEEL from calciner emissions were likely reduced in the soil from the emitted divalent Hg(II) species (known to be the primary form in the stack gas and readily deposit close to the source) to elemental Hg(0) where it was subsequently reemitted to the atmosphere. Once reemitted, the Hg(0) would become part of the global Hg pool which does not readily deposit locally (see Reference 1). However, this reemitted Hg does contribute to the increasing global atmospheric Hg pool, a fraction of which eventually is oxidized to Hg(II) and then subsequently deposited elsewhere. Confirmation and quantification of snow and soil Hg reemission losses will be pursued using flux chamber measurements in FY 2001.

ACCOMPLISHMENTS

Samples of snow (n = 130), rain (n = 6), soil (n = 103), and air (n = 5) were successfully collected on the INEEL and at 11 regional background sites from January through March 2000. Snow samples were successfully obtained from the Upper Fremont Glacier, Wyoming, in May (n = 20) and the Inilchek Glacier, Central Asia, in July (n = 50). This sampling required development and successful implementation of ultraclean sampling techniques necessary for consistent preservation of ambient Hg at parts-per-trillion levels, and successful and safe field operations in harsh winter conditions, even at some high-altitude locations. All samples except one passed rigorous laboratory QA/QC procedures, which included duplicate analyses on every sample (<10% variation), spike recoveries and quality control check samples every 10 samples ($\pm 10\%$), and bubbler blanks for background correction.

The major conclusions from this field research are as follows:

- Mercury concentrations in precipitation fallout are very low both on the INEEL and regionally. The highest concentration found on the INEEL during calciner operations was 141 ng/L, less than one-tenth the EPA's MCL for Hg in drinking water (2 ppb or 2,000 ng/L). Most concentrations on the INEEL were significantly lower. Concentrations at regional background sites were much lower and similar to the background fallout rate (3 ng/L) expected for this region. This background fallout rate is as low or lower than anywhere else in the U.S.
- Most of the Hg in the calciner emissions, which is almost 100% Hg(II), is deposited within 10 km of the stack, based on reconnaissance field sampling and model validation work done in 1999.¹⁰
- Mercury concentrations in surface soil near INTEC (15 to 20 ng/g) increase slightly at distances closer than 5 km, indicating a contribution from the calciner. However, the concentrations measured at all locations near INTEC are very low compared background Hg concentrations in similar soils around the U.S. (50 to 70 ng/g).
- A mass balance of the soil sampling data near INTEC indicate 40 to 300 kg of Hg from calciner emissions remain in the surface soil. This is estimated to be 3 to 20% of the cumulative 37 years of Hg deposition from the calciner (est. 1,500 kg).
- It is likely that most of the Hg fallout from calciner operations was initially deposited as Hg(II) close to INTEC, subsequently reduced in the soil to Hg(0), and then reemitted to the atmosphere where it would have become part of the global atmospheric Hg pool. Elemental Hg(0) does not readily deposit to the ground, and may be transported long distances in the atmosphere before a small fraction of it is oxidized and subsequently deposited on the ground. Confirmation and quantification of snow and soil Hg reemission losses on the INEEL will be pursued using flux chamber measurements in FY 2001.

Future Work

Future work includes continued precipitation and soil sampling on the INEEL and regionally, and new efforts on reemission flux measurements and aquatic ecosystem sediment sampling.

To better understand Hg cycling on a global scale, additional parameters affecting Hg speciation and transport between various environmental compartments (air, soil, sediment, and biota) may need to be

investigated. One area currently thought to be important, but of which very little is known, is the effect of soil microorganisms and biochemistry.

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